Document made available under the Patent Cooperation Treaty (PCT)

International application number: PCT/US05/009308

International filing date: 16 March 2005 (16.03.2005)

Document type: Certified copy of priority document

Document details: Country/Office: US

Number: 60/553,390

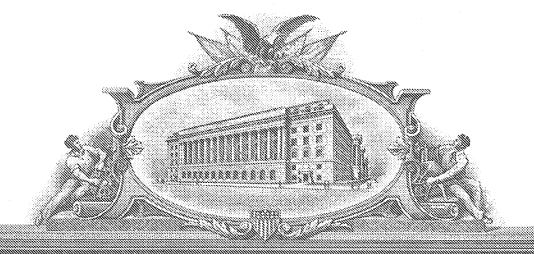
Filing date: 16 March 2004 (16.03.2004)

Date of receipt at the International Bureau: 12 May 2005 (12.05.2005)

Remark: Priority document submitted or transmitted to the International Bureau in

compliance with Rule 17.1(a) or (b)





'40) And 40) vardh andse, pressents, suam, (cones;

UNITED STATES DEPARTMENT OF COMMERCE

United States Patent and Trademark Office

May 04, 2005

THIS IS TO CERTIFY THAT ANNEXED HERETO IS A TRUE COPY FROM THE RECORDS OF THE UNITED STATES PATENT AND TRADEMARK OFFICE OF THOSE PAPERS OF THE BELOW IDENTIFIED PATENT APPLICATION THAT MET THE REQUIREMENTS TO BE GRANTED A FILING DATE.

APPLICATION NUMBER: 60/553,390

FILING DATE: March 16, 2004

RELATED PCT APPLICATION NUMBER: PCT/US05/09308

1316978

Certified by

Under Secretary of Commerce for Intellectual Property and Director of the United States Patent and Trademark Office

PTO/SB/16 (01-04) Approved for use through 07/31/2006. OMB 0651-0032

U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

PROVISIONAL APPLICATION FOR PATENT COVER SHEET

This is a request for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

Express Mail Label No. EV240599115US

		INVENTO	R(S)			
Given Name (first and middle [if any])		Family Name or Surname		(City s	Residence (City and either State or Foreign Country)	
Craig P.		Jacobson			Moraga, CA	
Lutgard C.		De Jonghe		Lafayet		
Additional inventors are being	a named on the					,
Additional inventors are being named on theseparately numbered sheets attached hereto TITLE OF THE INVENTION (500 characters max)						0
Compact Fuel Cell with Edge Current Collection						ഗന്
						5.5°C
Direct all correspondence to: CORRESPONDENCE ADDRESS						5535 60/8
Customer Number: 08076						1556 60
OR						<u> </u>
Firm or Individual Name						
Address						
Address		_	_			
City			State		Zip	*
Country			Telephone		Fax	
	ENCLOS	ED APPLICATION PAI	RTS (check	all that apply)		
X Specification Number of Pages 8 CD(s), Number						
X Drawing(s)Number of Sheets 5 Other (specify)						
Application Data Sheet.	See 37 CFR 1.76			., ,,		
METHOD OF PAYMENT OF			PLICATION F	ORPATENT		- -
Y Applicant claims arrell antity status Con 27 OFD 4 07						
Amount (\$)						
A check or money order is enclosed to cover the filing fees.						
The Director is hereby authorized to charge filing fees or credit any overpayment to Deposit Account Number: 12-0690 80.00						
Payment by credit card. Form PTO-2038 is attached.						
The invention was made by a United States Government.			under a contr	ract with an agenc	y of the	
No.						
	.S. Government a	gency and the Government	contract numb	herare: U.S. Da	enartment of Fnero	v Drima
Contract No. DE-A				DCT 410. 0.D. DC	parament of Energ	y, Finite
		[Page 1 o	f1]	<u>, , , , , , , , , , , , , , , , , , , </u>	· lalou	
Respectfully submitted,		•••				
SIGNATURE / / / // //				REGISTRATION NO. 46,470 (If appropriate)		
TYPED or PRINTED NAME Charles R. Nold				Docket Number	IB-1942P	

TELEPHONE 510-486-6503 USE ONLY FOR FILING A PROVISIONAL APPLICATION FOR PATENT

This collection of information is required by 37 CFR 1.51. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 8 hours to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Mail Stop Provisional Application, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.

COMPACT FUEL CELL WITH EDGE CURRENT COLLECTION

Inventors: Craig P. Jacobson

Residence: Moraga, CA

Lutgard C. De Jonghe

Residence: Lafayette, CA

CROSS-REFERENCE TO RELATED APPLICATIONS

[001] Not applicable

STATEMENT OF GOVERNMENTAL SUPPORT

[002] This invention was made during work supported by U.S. Department of Energy under Contract No. DE-AC03-76SF00098. The government has certain rights in this invention.

BACKGROUND OF THE INVENTION

FIELD OF THE INVENTION

[003] The present invention relates to the field of electrochemical devices, and more specifically solid oxide fuel cells, SOFC.

RELATED ART

[004] Steadily increasing demand for power and the atmospheric build up of greenhouse and other combustion gases has spurred the development of alternative energy sources for the production of electricity. Fuel cells, for example, hold the promise of an efficient, low pollution technology for generating electricity. Because there is no combustion of fuel involved in the process, fuel cells do not create any of the pollutants that are commonly produced in the conventional generation of electricity by boilers or furnaces and steam driven turbines.

[005] Unfortunately, the present cost of electrical energy production from fuel cells is several times higher than the cost of the same electrical production from fossil fuels. The high cost of capitalization and operation per kilowatt of electricity produced has delayed the commercial introduction of fuel cell generation systems.

[006] A conventional fuel cell is an electrochemical device that converts chemical energy from a chemical reaction with the fuel directly into electrical energy. Electricity is generated in a fuel cell through the electrochemical reaction that occurs between a fuel (typically hydrogen produced from reformed methane) and an oxidant (typically oxygen in air). This net electrochemical reaction involves charge transfer steps that occur at the interface between the ionically-conductive electrolyte membrane, the electronically-conductive electrode and the vapor phase of the fuel or oxygen. Water, heat and electricity are the only products of one type of fuel cell system designed to use hydrogen gas as fuel. Other types of fuel cells that have been developed include molten carbonate fuel cells, phosphoric acid fuel cells, alkaline fuel cells, proton exchange membrane fuel cells. Because fuel cells rely on electrochemical rather than thermo-mechanical processes in the conversion of fuel into electricity, the fuel cell is not limited by the Carnot efficiency experienced by conventional mechanical generators.

[007] Solid-state electrochemical devices are normally cells that include two porous electrodes, the anode and the cathode, and a dense solid electrolyte membrane disposed between the electrodes. In the case of a typical solid oxide fuel cell, the anode is exposed to fuel and the cathode is exposed to an oxidant in separate closed systems to avoid any mixing of the fuel and oxidants due to the exothermic reactions that can take place with hydrogen fuel.

[008] The electrolyte membrane is normally composed of a ceramic oxygen ion conductor in solid oxide fuel cell applications. In other implementations, such as gas separation devices, the

solid membrane may be composed of a mixed ionic electronic conducting material ("MIEC").

The porous anode may be a layer of a ceramic, a metal or, most commonly, a ceramic-metal composite ("cermet") that is in contact with the electrolyte membrane on the fuel side of the cell.

The porous cathode is typically a layer of a mixed ionically and electronically-conductive (MIEC) metal oxide or a mixture of an electronically conductive metal oxide (or MIEC metal oxide) and an ionically conductive metal oxide.

[009] Solid oxide fuel cells normally operate at temperatures between about 900°C and about 1000°C to maximize the ionic conductivity of the electrolyte membrane. At appropriate temperatures the oxygen ions easily migrate through the crystal lattice of the electrolyte. However, most metals are not stable at the high operating temperatures and oxidizing environment of conventional fuel cells and become converted to brittle metal oxides. Accordingly, solid-state electrochemical devices have conventionally been constructed of heat-tolerant ceramic materials. However, these materials tend to be expensive and still have a limited life in high temperature and high oxidation conditions. In addition, the materials used must have certain chemical, thermal and physical characteristics to avoid delamination due to thermal stresses, fuel or oxidant infiltration across the electrolyte and similar problems during the production and operation of the cells.

[010] Since each fuel cell generates a relatively small voltage, several fuel cells may be associated to increase the capacity of the system. Such arrays or stacks generally have a tubular or planar design. Planar designs typically have a planar anode-electrolyte-cathode deposited on a conductive interconnect and stacked in series. However, planar designs are generally recognized as having significant safety and reliability concerns due to the complexity of sealing of the units and manifolding a planar stack.

[011] In addition, conventional stacks of planar fuel cells operated at the higher temperature of approximately 1000°C have relatively thick electrolyte layers compared to the porous anode and cathode layers applied to either side of the electrolyte and provides structural support to the cell. However, in order to reduce the operating temperature to less than 800°C, the thickness of the electrolyte layer has been reduced from more than 50-500 microns to approximately 5-50 microns. The thin electrolyte layer in this configuration is not a load bearing layer. Rather, the relatively weak porous anode and cathode layers must bear the load for the cell. Stacks of planar fuel cells supported by weak anodes or cathodes may be prone to collapse under the load.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG 1 shows a graph depicting the performance of CSO-electrolyte SOFC membrane at various temperatures, with H_2 - H_2 O/air

- FIG 2 shows membrane architecture
- FIG 3 shows Unit cell, view A
- FIG 4 shows Unit Cell, view B
- FIG 5 shows series stacking arrangement of the unit cells
- FIG 6 Relation between achievable power densities, area specific power density, and fuel cell membrane dimensions, at fixed aspect ratio

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENT

Solid oxide fuel cells offer the potential of high volumetric power density combined with fuel flexibility. Considerable progress is being made in raising the performance of solid oxide fuel cells, and as an example, the present inventors were the first to demonstrate that power densities of as much as $2W/cm^2$ could be obtained for supported thin-film yttria -stabilized

zirconia (YSZ) solid oxide fuel cells, at 800°C.¹ While this result was encouraging, further reductions in temperature are necessary. Such reduction in operating temperature on the one hand makes the use of metallic interconnects and support electrodes possible, allowing for vast cost reduction, but on the other hand requires new ways of configuring fuel cells so that current can be collect with minimal resistive loss. The present invention contemplates a stack temperature regime for semi-portable or fully portable solid oxide fuel cells is between 600 and 650°C.

Several materials choices impose themselves as suitable for the present invention. A first one is the choice of the solid electrolyte. For the present purpose, preferred Samaria-doped ceria (CSO), with a composition of Ce_{0.8}Sm_{0.15}O₂. Previous work by the present inventors has demonstrated obtainable area specific power in excess of 500mW/cm² between 600 and 650°C, for a fuel cell membrane with a Lanthanum-Strontium-Cobalt-Nickel oxide (LSCN)/CGO (Gadolina-doped Ceria) composite cathode, and a Ni/CSO composite anode.² Test results on this system are shown in FIG 1.

The present invention contemplates an alternative anode and cathode that may to lead to performances exceeding those shown in Fig 1, additionally allowing for efficient edge current-collection within defined dimensional limits.

The devices of this invention are contemplated to have at least 200 mW/cm2 at 600-650oC for a unit cell solid oxide fuel cell. The SOFC stack has projected power densities ranging from 0.8 kW/liter (@200mW/cm2) to 1.75 kW/liter (@400mW/cm2), and can be assembled simply by combining the unit cells, without introducing significant additional sealing

5

¹ S. de Souza, S.J. Visco, and L.C. De Jonghe, "Reduced-temperature solid oxide fuel cell based on YSZ thin-film electrolyte," *J. Electrochem. Soc.*, **144**, L35-L37 (1997).

² C.P. Jacobson, S.J. Visco, L. De Jonghe, unpublished results, 1999.

or manifold difficulties. The invention contemplates that this performance will be achieved with fuel/oxidant combinations of (H₂, H₂O)/air and of simulated reformer gas/air.

The fuel cells may also be run on other fuels, such as alcohols.

Technical Description: Membrane Structure

A structure in accordance with one embodiment of this invention is the membrane structure is shown in FIG 2. The fuel electrode consists of a porous ferritic stainless steel³, 0.4 mm thick, activated by incorporation of a Ni/CSG dispersion. Additionally, stable increased catalytic activity may be obtained by post-infiltration with compounds that form nano-scale catalyst particles near or at the electrolyte/ anode interface.⁴ The ferritic steels have thermal expansion coefficients that can match those of the ceramic electrolyte nearly perfectly, thereby avoiding thermal stresses and allowing for high heating rates and thermal cycling. These structural concepts are illustrated in FIG 2. The cathode current collection needs to be facilitated by a supporting stainless steel mesh that is incorporated in the LSCN cathode, as also shown in FIG 2. The cathode-supported thin film CSO is produced by colloidal processing and co-firing⁵ in reducing atmospheres, and has already been shown to be possible in our associated LBNL programs. Strongly reduced sintering temperatures of the CSO electrolyte, well below 1000°C have been achieved by the present inventors.⁶

Unit Cell Architecture

The membrane is mounted in a stainless steel supporting structure that in part also serves to provide a current path for series/parallel operation, when incorporating the proper combination

³ Steven J. Visco, Craig P. Jacobson, Igor Villareal*, Andy Leming, Yuriy Matus and Lutgard C. De Jonghe, DEVELOPMENT OF LOW-COST ALLOY SUPPORTED SOFCS, Proc. ECS meeting, Paris, May 2003.

⁴ K. Yamahara, Craig P. Jacobson, Steven J. Visco, Lutgard C. De Jonghe, High-Performance Thin Film SOFCs for Reduced Temperature, Proceedings SSI 14, Monterey, CA, 2003.

S.J. Visco, C.P. Jacobson, L.C. De Jonghe, U.S. Pat. No. 6458170, October 1 (2002).

and placement of insulating and conducting sealing compounds. The general design is illustrated in FIGS 3 and 4. This invention contemplates that unit cells are be prepared for which the active cell area is approximately 4 cm². It should be noted that a complete unit cell incorporates all the challenges associated with combining such cells in a series stack arrangement.

Series Stacking

Series stacking of unit cells can be achieved as shown in FIG 5. In this figure, the placement of the insulating and conducting sealing compounds, as well as he current path is indicated.

Dimensional and Performance Considerations

The dimensions of the unit cells are constrained by the need to have efficient edge current-collection. This is in turn determined by the in-plane conductivity of the electrodes, by non-active edge areas, etc. Calculations based on these factors and on the known electronic resistance of the various materials involved in the electrodes indicate that a maximum length for edge current collection, with a potential drop of less than 50 mV, is between 4 and 5 cm. The projected performance of the fuel cell will therefore be sensitive to a number of geometrical factors as well as to the intrinsic power per unit electrode area. FIG 6 considers these factors, with an example fuel cell membrane aspect ratioo of 2. It follows that impressive volumetric power densities are possible within the indicated design working range. Even at modest area specific power densities of 200mW/cm², 1kW/liter can be achieved. More optimistic, but by no means unreasonable, assumptions about achievable area specific power densities puts the stack volumetric power density at an extraordinary power density of well over 2 kW/liter.

⁶ M. Cruz, C.P. Jacobson, S.J. Visco, and L. De Jonghe, unpublished results. (2003)

Attorney Docket: IB-1942P Lawrence Berkeley National Laboratory

The metal construction of the cell housing and the stack avoid stress transfer to the membranes, so that compression sealing can be used for cell sealing and stack manifolding. The flexibility in the unit cell architecture and the series assemblies can be readily envisioned to lead to combinations that range from a few 10s of Watts to 10s of kilowatts, in highly compact power generating devices.

[012] All references, patents, patent applications disclosed herein are expressly incorporated by reference in their entirety for all purposes.

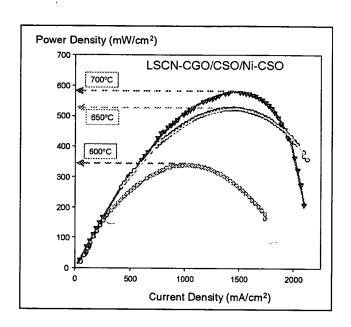


FIG 1

Compact Fuel Cell Architecture- Membrane structure

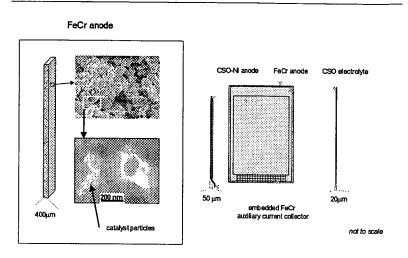
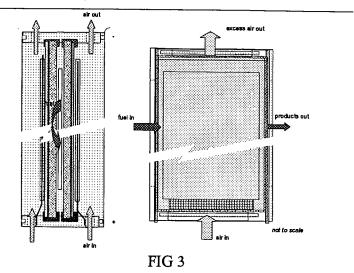


FIG 2

Compact Fuel Cell Architecture- unit cell



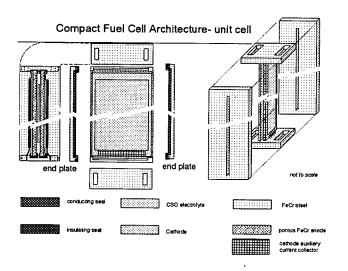


FIG 4

Compact Fuel Cell Architecture- series stacking

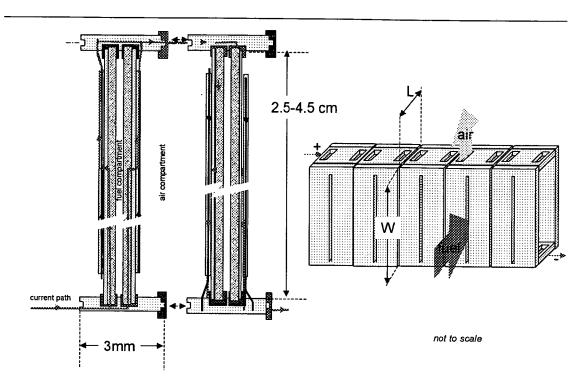


FIG 5

Compact Fuel Cell Architecture- dimensional considerations

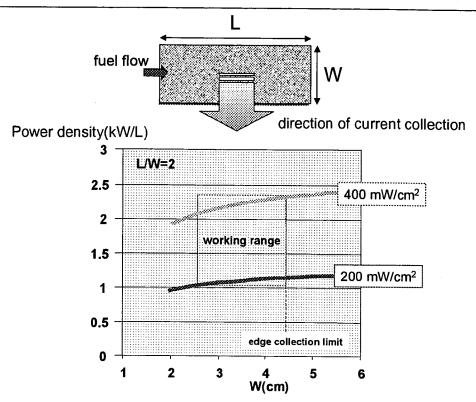


FIG 6